Aerosol formation over the Boreal forest in Hyytiälä, Finland: monthly frequency and annual cycles ? the roles of air mass characteristics and synoptic scale meteorology

E. D. Nilsson, M. Kulmala

Atmos. Chem. Phys. Discuss., 6, 10425–10462, 2006
www.atmos-chem-phys-discuss.net/6/10425/2006/

Atmospheric Chemistry and Physics Discussions

Aerosol formation over the Boreal forest in Hyytiälä, Finland: monthly frequency and annual cycles – the roles of air mass characteristics and synoptic scale meteorology

E. D. Nilsson[1] **and M. Kulmala**[2]

[1]Stockholm University, Department of Applied Environmental Science, 10691 Stockholm, Sweden
[2]University of Helsinki, Department of Physical Sciences, P.O. Box 64, 00014 Helsinki, Finland

Received: 29 September 2006 – Accepted: 9 October 2006 – Published: 18 October 2006

Correspondence to: E. D. Nilsson (douglas.nilsson@itm.su.se)

Abstract

New atmospheric particles with diameters of 3–10 nm and their subsequent growth to cloud condensation nucleus have been observed at various places in the European boundary layer. These events have been observed simultaneously within wide geographical areas (over 1000 km) in connection to specific weather systems, the cold air behind cyclones. Here we show that atmospheric aerosol formation (i.e. nucleation and initial growth) is favoured by the outbreak of cold Arctic air over northern Europe. Aerosol formation was about twice as common in Arctic air as in sub-Polar air, and even more so compared to other air masses. The most important general factor favouring aerosol formation in Arctic air and marine air was weaker competing condensational sink (CS) for the precursor gases (less pre-existing aerosols), while high CS prevented aerosol formation in heated sub-Polar air and mid-latitude air. High SO_2 levels favoured nucleation in continental air and high UV-B radiation in sub-tropical air. The critical factor that determined if aerosol formation would start on a day with Arctic air was the UV-B radiation. The same applied to sub-Polar air and continental air, while increased SO_2 concentration could trigger formation in heated sub-Polar and mid-latitude air, and reduced CS could cause formation in mid-latitude, marine or mixed/transient air. We speculate that strong emissions of volatile organic compounds from the Boreal forest and strong boundary layer dynamics may have caused aerosol formation in sub-Polar air masses and air in transition from a marine to a continental character. The monthly frequency of Arctic air masses and the probability for photo-chemically driven aerosol formation explains the observed annual cycle in monthly particle formation frequency as well as much of the inter annual variability. The same cyclones that transport cold, clean air from the Arctic to Europe will also transport warm polluted air in the other direction, which help cause the Arctic Haze phenomena. The cyclones have a key role for the atmospheric aerosol life cycle in mid to high latitudes. Due to the observed growth to the size of CCN in one to two days, there is a potential feed back from the effects on the CCN population and cloud albedo even within the same weather system,

ACPD

6, 10425–10462, 2006

but also on the climatic time scale.

1 Introduction

The importance of atmospheric aerosols to the global radiation balance, to cloud formation, and to alleged human health effects has motivated several recent studies on dynamics of atmospheric aerosols. Formation of new nm sized aerosol particles, nucleation, and subsequent growth to the size of cloud condensation nuclei (CCN) has been observed in various sites around the world (Kulmala et al., 2004a), e.g. in the continental boundary layer all over Europe (Mäkelä et al., 1997; Kulmala et al., 2001a; Birmilli and Wiedensohler, 2000; Coe et al., 2000; Väkevä et al., 2000; Held et al., 2004; Rodriguez et al., 2005) and recently also during the Asian Pacific Regional Aerosol Characterization Experiment (McNaughton et al., 2004; Buzorius et al., 2004), eastern USA (Gaydos et al., 2005) and an Indian mega-city (Mönkkönen et al., 2005). The longest time sequence of observations is from Hyytiälä (61°51′ N 24°17′ E) in Finland, where measurements with a differential mobility particle sizer (DMPS, measures the aerosol number concentration from 3–610 nm dry diameter) begun in January 1996. From January 1996 to June 2000 there were 287 days with aerosol formation (including the two most well defined categories of aerosol formation events according to Mäkelä et al. (2000). The particle formation is usually observed in the late morning as the appearance of 3 nm particles at the lower size range of the Differential Mobility Particle Sizer (DMPS), see Fig. 1 for a typical example, followed by growth towards CCN size. The onset of aerosol formation is very well correlated with the onset of strong turbulence, as observed by a SODAR (Nilsson et al., 2001a). It was better correlated with the onset of turbulence than with the rapid increase in UV-B in the morning hours. Also other investigators have found a relation between boundary layer dynamics and the nucleation onset (Stratmann et al., 2003; Uhrner et el., 2003; McNaughton et al., 2004; Gaydos et al., 2005). There are several possible explanations to why boundary layer dynamics could trigger aerosol formation, e.g. intermittent mixing over gradients

ACPD
6, 10425–10462, 2006

(Nilsson and Kulmala, 1998), atmospheric waves (Nilsson et al., 2000) or turbulent fluctuations (Lauros et al., 2006). Detailed boundary layer models with aerosol dynamics has only begun to connect these theoretical possibilities with the typical observations in Hyytiälä (Hellmuth, 2006a, b).

It has been found that the aerosol formation is favoured by the outbreak of Arctic air, sometimes Polar air, over northern Europe (Nilsson et al., 2001b). Under these synoptic conditions, the aerosol formation was observed over a distance of 1000 km from Värriö in northern Finland (67°48′ N 29°30′ E), over Hyytiälä, to Takhuse in Estonia (58°31′ N 24°56′ E), when all of these stations were in the same air mass. The sequences of days with aerosol formation started and ended as the synoptic weather changed and with the passage of the Arctic and Polar fronts. Connection between large-scale atmospheric motions and aerosol formation has been found also in other regions and climate zones. Birmilli and Wiedensohler (2000) related aerosol formation in central Europe to large-scale subsidence from the Alps. McNaughton et al. (2004) found enhanced nucleation in post-frontal air over the Chinese Sea on a scale of 200 000 km^2, or over a distance of 400 km (Buzorius et al., 2004). While we for Hyytiälä found a preference for aerosol formation in air in transition from Arctic marine conditions towards continental conditions, McNaughton et al. found aerosol formation in previously continental air that was in transition over the sea, gradually forming a marine boundary layer. Vana et al. (2004) extended the Nilsson et al. (2001b) study to include 157 days of measurements from the same three stations, to find that aerosol formation extended over 1000 km in 18 days. The intention of the current work is to extend the analysis in time rather than space, to study how aerosol formation in Hyytiälä relates to synoptic weather during a period long enough to include several annual cycles: the period January 1996 to June 2000.

2 Methods

2.1 Air mass analysis

Berliner Wetterkarte (Institute für Meteorologie, 1997) provides a classification scheme for air masses on daily basis at 00:00 UTC. The air mass classification is based on back trajectories and typical thermodynamical properties. The air mass may be characterised by class and character. Based on this definition the air masses are divided in Arctic (A), sub-Polar (P) and heated sub-Polar (Ps), Subtropical (S), middle latitude (Sp), and tropical air mass classes (T). The influence from sea or continents, on energy content (heat and water vapour), is responsible for much of the formation of respectively air mass and for transitions in between them, and for the extent of continental or marine influence. In this manner each air mass may be characterized as continental (c), marine (m) or mixed air in transition (x). For example, Arctic air may thus be defined as mA, xA and cA. We have used the air mass classification and surface front analysis made daily for 00:00 UTC (02:00 local Finnish wintertime). We have established the types of air masses that were located over the experimental site, the presence of fronts, their passages over Hyytiälä and if there where conditions of warm or cold air advection behind cold fronts, especially so called cold air outbreaks. In addition, we have located the latitudinal approximate surface position of the Arctic front and polar fronts along the 25° East longitude line (which runs close to Hyytiälä).

2.2 DMPS measurements and classification of aerosol formation days

The dry aerosol number size distributions were measured with a Differential Mobility Particle Sizer (DMPS) system in 10 min cycles at 2 m height in Hyytiälä, which gives a continuous view of the distribution and evolution of the aerosol particles. The DMPS system used here consists of two DMPS systems. The first one includes a TSI 3025 UFCPC and a Hauke-type short DMA (Differential Mobility Analyzer). It measures particles between 3 and 20 nm in dry diameter. The second one includes a TSI3010

ACPD
6, 10425–10462, 2006

CPC and a Hauke-type medium DMA and measures particles between 20 and 500 nm. Aalto et al. (2001) describes this system in more details, together with other aerosol physical measurements.

The aerosol formation events days were classified by Mäkelä et al. (2000) and updated by dal Maso et al. (2005) in four categories according to the Hyytiälä DMPS measurements. 1) Cases that showed a clear nucleation mode that was easily distinguishable until it had grown to at least the Aitken mode. 2) There were fewer particles formed, or some background concentration existed in the smallest DMPS channels, or the growth was less nice, than for class 1. 3) Same problems as for class 2, but much worse. In this class it was difficult to see the nucleation mode at times. We will only consider class 1 and 2 as clear aerosol formation days in our analysis.

2.3 Supporting chemical and meteorological measurements

A wide range of local meteorological parameters and concentrations of trace gases are measured at SMEAR II station continuously (see Kulmala et al., 2001a). In the present study we have utilized SO_2, and UV-B data. The SO_2 is measured using the fluorescence technique (TEI 43 BS, Thermo Environmental) and UV-B using a pyranometer (Solar Light SL 501A).

3 Results

3.1 Aerosol formation frequency

The first part of the database that we will take a look at is the daily classification of aerosol formation, or absence of aerosol formation, from which we have calculated monthly frequencies of days with aerosol formation. Although with substantial inter annual variability, a clear annual pattern can be seen in the monthly occurrence of aerosol formation days, see Fig. 2. Most pronounced, and present every year, is the

spring peak. Between 1/3 and 2/3 of the days in the spring are aerosol formation days, but the peak month shifts from April (in 1996–1998) to May (in 1999–2000). The spring maximum in aerosol formation is surrounded by winter and summer minima. Finally, there is a small autumn peak (September or October). This annual cycle was observed already in the first year of data (Mäkelä et al., 1997) and has been described in detail by Dal Maso et al. (2005), but not in the context of synoptic weather and annual cycles in meteorology.

3.2 Air masses over different seasons

The second part of the database is the daily classification of the air mass type over Hyytiälä, from which we have calculated monthly frequencies of aerosol formation. The monthly average frequency of each air mass type occurring at Hyytiälä is shown in Fig. 3. Sub-Polar (P) air masses, the most common on average, varies between 30 and 45% without any obvious annual cycle. The 2nd most common air mass, the Arctic (A) air has a pronounced annual cycle. It is almost entirely absent during the summer months of June, July and August, when most of the Arctic is not cold enough for A air masses to form. Instead the formation region of P air is extended northward. During the summer months, Ps, Sp and S air masses increase instead. The strongest seasonal variability in A air was found in mixed air (xA), which largely is air in transition from marine (mA) to continental air (cA), during cold air out breaks, which also has a minimum in summer. The xA air masses return in the autumn, remain common throughout winter and spring and disappear again in the early summer, see Fig. 4.

3.3 Annual cycles in aerosol formation and air mass and the influence of the position of the Arctic front

We have further combined the monthly frequencies of aerosol formation days and air mass types. Looking at the distribution of different air masses exclusively on days with aerosol formation we can see that more than half of the aerosol formation days oc-

ACPD
6, 10425–10462, 2006

curred in Arctic air, with sub-Polar air as the 2nd most common air mass, see Fig. 5. Aerosol formation is rare in the remaining air mass types. Even if we consider that Arctic and sub-Polar air dominates at Hyytiälä, except in the summer, due to its northern position, there is still a preference for aerosol formation in Arctic air.

The Arctic front constitutes the border between the Arctic and sub-Polar air masses. Figure 6 shows the monthly average position of the Arctic front along the 25° E Longitude, which runs almost through Hyytiälä. If we recall that Hyytiälä is located near Latitude 64° N, it is clear that the Arctic front on average is located in the vicinity of Hyytiälä throughout autumn and winter, and even a bit south of it during spring, while in summer it is located further north, or in most cases even absent. Cyclones and their associated warm fronts and cold fronts can be considered to move along the average Arctic front and cause variability in the momentary front position on a the synoptic time scale (~5 days). Therefore, an average position of the Arctic front near Hyytiälä implies that Hyytiälä will during that month experience many front passages and postfrontal cold air out breaks, such as those observed by Nilsson et al. (2001b). The frequency of occurrence of various air masses is a function of the season, obviously, but should also be a function of the position of the measurement station. Therefore, a station further north or south will have a different position to the average front and experience a different annual cycle in the air mass characteristics. From this follows that if there is a link between air mass and aerosol formation, the annual cycle of aerosol formation frequency will also change with the geographic position.

3.4 Air mass characteristics

Table 1 summarises the average conditions for some key factors that characterise each air mass type. We have included SO_2 and UV-B as representing the precursor production. At least they are relevant if sulphuric acid participates in the aerosol formation as supported by recent observations (e.g. Kulmala et al., 2004a, 2006). UV-B may also have a more general relevance if precursors form from photochemical disintegration of terpenes or other primary organic emissions from the Boreal forest. There are ob-

ACPD
6, 10425–10462, 2006

viously many other parameters that one would like to include in this comparison, but they are not available in Hyytiälä as long-term measurements (e.g. neither NH_3, OH, nor volatile organic emissions). For UV-B, we have averaged only the period 09:00–12:00 h local time, during which a majority of the aerosol formation events start, and during which UV-B typically experience a rapid increase due to the rising sun. To represent the competing sink term, we use the Condensational Sink (CS), see for example Kulmala et al. (2001b), for definition, based on the Hyytiälä DMPS measurements. Note that the CS here does not include particles >610 nm D_p, or the effect of water uptake, since hygroscopicity is not available, except on campaign basis. Therefore, it is a lower estimate.

All air masses describe an average diurnal cycle with a reduction in CS during the morning. This is in agreement with the reduction in both Aitken and accumulation mode on the order of 50% prior to the first observation of new particles that was demonstrated by Nilsson et al. (2001a). We explained this with entrainment of cleaner air from aloft driven by the rapid development of the mixed layer in the morning. About 1/3 of the aerosol formation days had this behaviour, and it may help initiate aerosol formation on those days (see also Hellmuth, 2006a, b). On the other hand, especially in A, P and Ps air masses, one can see a recovery of the CS after typically 15 local time in the average afternoon due to growth of the new nucleation mode into an Aitken mode and towards CCN size. Diurnal cycles are not the main focus of this study, but one have to realise how dependent CS is on the time of the day, before comparing CS in different air masses etc. The contribution of the new aerosol to the CS will move the CS away from the value that are representative of the air mass. Because of this, we have chosen to compare both the full days, the period 09:00–12:00 during which the aerosol formation usually begin, and the 3-h long period before that, see Table 2. Even in the average CS in Table 2 we can see the effect of dilution by entrainment since the CS from 09:00–12:00 is systematically lower than the CS from 06:00–09:00, except for Sp air without nucleation. In the following we will focus less on the full day average CS, for the reasons given above, but these are still included in the tables to allow comparison

to other studies.

3.4.1 Source proxies: UV-B and SO_2

If we first take a look at the respectively average properties for each air mass type over all days, we can see that those air masses that mainly exist during the summer have higher average incoming UV-B radiation, ~0.6 Wm^{-2}, or even more for S, while A and P air has ~0.3 Wm^{-2}, presumably due to the lower solar angle in autumn, winter and spring. If we compare aerosol formation days with the average days, we can see a distinctly higher incoming UV-B radiation in A and P air, ~40%. Although these air masses are generally more related to aerosol formation than other air masses, there is obviously a subset of them with a higher average UV-B during days with aerosol formation that reach about the same UV-B level as in other air masses, and this subset is associated with the aerosol formation days. Cold air advection from the north is in general associated with clear skies and with only a partial or broken cloud cover of "fair weather clouds" (cumulus mediocris and humilis), but the statistics in Table 1 suggest that it is even in this group important with relative differences in cloudiness and photochemistry.

One would perhaps expect lower concentrations of SO_2 in the air masses that originate from the north, but there is small or insignificant difference in SO_2 between the different air masses in general. Arctic air actually has the highest average SO_2 concentration. It may be less strange than it sounds. Sulphur dioxide has a relatively short lifetime, ~1 day in the summer Arctic (e.g. Nilsson and Leck, 2002), and cannot originate as far away as the more densely populated central or eastern Europe even if this was a formation region for Arctic air (which it is not). Nor can it have a generally natural origin in the formation regions of P and A air, the Arctic Ocean, as an oxidation product of Dimethylsulfide, since these waters are mainly ice covered in winter and spring. Perhaps if the ice break up close to land followed by an algae bloom in the late spring their could be a contribution. However, marine air has the lowest average SO_2 concentration (0.25 ppb) of all categories in Table 1, so even if DMS may contribute

ACPD
6, 10425–10462, 2006

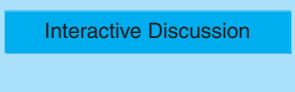
occasionally, it cannot be the general source for SO_2. On the other hand, emissions from biomass (wood) and fossil fuel burning for heating is quite intensive in Northern Europe in winter and spring, and would be even more so each time cold air from north dwell over the region. Hence SO_2 are likely to originate from relatively local sources, and will not originate from the region as the air mass. Arctic air actually decrease its SO_2 concentration with on average 30%, when there is aerosol formation, while P air is on average unaffected, and Ps and Sp increase ~50–100% to about 1 ppb on average. This may indicate that when aerosol formation occurs in southern air masses, it is associated with increased SO_2 concentration somewhat approaching levels found in studies on aerosol formation in central Europe.

3.4.2 Sink proxy: condensational sink

In general, if we compare all full days with or without aerosol formation, the condensational sink is smaller in the A and P (also in the Ps) air masses, ~3–4×10^{-3} s^{-1}, while Sp from central and eastern Europe has an average value of almost 5×10^{-3} s^{-1}. If we take a look at the average CS values 06:00–09:00 and 09:00–12:00 local time, we see a more differential picture: the average CS increase from ~2×10^{-3} s^{-1} for A air to ~4×10^{-3} s^{-1} for P air over ~5×10^{-3} s^{-1} and ~6–7×10^{-3} s^{-1} for Sp air masses. The few cases of S air mass has CS values >10^{-2} s^{-1}. In the same way, when we look at the average CS during and before the onset of formation, we go from ~3×10^{-3} s^{-1} to ~5×10^{-3} s^{-1} over ~4×10^{-3} s^{-1} when going from marine to continental over mixed air. This is the background before we consider what characterise days with and without aerosol formation in a given air mass. The tables also confirm our assumption that full day CS averages would be obscured by the aerosol formation itself, as both the marine vs. continental character and the difference between days with and without aerosol formation appears to suffer from this problem.

The difference between CS on days with and without aerosol formation is least in P air, more significant in A and Ps air, and the largest difference, a factor ~3, is found in Sp air, see Table 2. The same picture is seen in Table 1 where the aerosol formation

ACPD
6, 10425–10462, 2006



days are compared to the average of all days. It appears that the pre-existing aerosol is important for the probability for aerosol formation: low CS favour aerosol formation, but not in the same way in all air masses. If we consider the marine vs. continental character of the air, we find an interesting pattern in the difference between days with and without aerosol formation. There is a large difference in marine air with almost half the average CS in days with aerosol formation, and about the same difference ($\sim 1.5 \times 10^{-3}$), also for mixed x air masses, but no significant difference for continental air, see Table 2. Considering that the study takes place in a transition zone where marine air from the oceans in the west and north are transferred towards a more continental character, this difference is interesting. We have seen previously that Arctic air favour nucleation, and we now see that while it is still preserving part of its marine character in the form of a lower CS, it continuous to favour aerosol formation, but the effect is lost when the transition is complete. It is also absent in the air that formed over the continents in the south and east from Hyytiälä, and in the average sub-Polar air.

4 Discussion

4.1 Key factors behind the monthly probability of aerosol formation

We have found that Arctic air is the most favourable air mass for aerosol formation and the key parameter appear to be low CS. However, at the first sight there is no similarity between the annual cycle of the Arctic air masses, or the Arctic front, and aerosol formation, besides the summer minimum. Clearly other factors will also influence the probability for aerosol formation, for example the rates of photochemical reactions related to the amount of solar radiation, or biogenic emissions of precursor vapours. As a proxy for such processes we will use a sinus formed function (near one in June, centred at mid-summer, and near zero in December centred at the mid-winter). It may represent the probability for photo-chemically driven aerosol formation, e.g. by the reaction of SO_2 and OH to form H_2SO_4 or formation of condensable organic vapours from pri-

ACPD
6, 10425–10462, 2006

mary emissions of volatile organic carbon or biogenic emissions related to the annual cycle in vegetation respiration or biological activity, see Fig. 7. UV-B measurements are not available for all years in the database, and there are frequent gaps in the data. However, the normalised annual average cycle of all available UV-B measurements between 09:00 and 12:00 h local time (the period when most aerosol formation events start) is very close to our proxy number in both phase and form. We have multiplied the frequency of Arctic air masses month by month with this proxy number.

The products resembles the observed average frequency of days with aerosol formation per average month very well, see Fig. 8. When we look at individual months, we can see that we manage to predict the spring maximum, the autumn maximum, the winter and summer minima, and part of the inter-annual variability in amplitude and phase, see Fig. 9. This suggest that the annual cycle in aerosol formation in Hyytiälä is caused by a combination of two different seasonal cycles: 1) the annual cycles in the synoptic weather and air mass transport and 2) the annual cycle in photochemistry or in the biologically driven emissions of organic precursors, both with an annual cycle that peak in the summer, or a combination thereof (see also Kulmala et al., 2004b).

4.2 Synoptic weather and the aerosol life cycle

The large spring peak in the aerosol formation and the decline in aerosol formation frequency with the arrival of the summer coincide with the late stage and the disappearance of the Arctic Haze phenomena. The Arctic Haze is caused by transport of anthropogenic aerosols and aerosol precursors into the Arctic, which in combination with weak aerosol sinks and dispersion within the central Arctic, creates an aged aerosol that remain airborne at high concentrations. Models and data analysis, e.g. Rahn (1982) and Heintzenberg and Larssen (1983), respectively, have shown that the Arctic Haze aerosol originates (at least in the European Arctic sector) mostly from European/Eurasian sources. The spring peak in Arctic Haze is partly a result of increased northbound transport due to increased cyclone activity over Europe in the spring, where each cyclone transport warm and polluted air into the Arctic, see Fig. 10.

ACPD
6, 10425–10462, 2006

We have learned in the current study that each cyclone, in particularly in the spring, also transports cold and clean Arctic air southward to Europe, which creates the right conditions for aerosol formation. It is accurate to, at least conceptually, identify the cyclones as the synoptic weather system which connects the aerosol source regions in Europe to the aerosol sink regions in the high Arctic, not only in one direction, as previously known for Arctic Haze, but bi-directional. Cyclones act in this system both to transport the aerosol, and to trigger aerosol formation (see Fig. 10). This should apply also to other regions with similar synoptic weather at similar latitudes, e.g. the USA/Canada, and perhaps also elsewhere. This is also consistent with the recent work by Tunved et al. (2005), based on five stations with DMPS measurements in Northern Europe (including Hyytiälä) and air mass and trajectory analysis: northward transport is associated with high aerosol concentrations of central European origin and gradual reduction in the concentrations due to dry and wet sinks, while southward transport from the Arctic is associated with low aerosol concentrations and gradual formation of new nucleation mode aerosol particles.

It is widely accepted that large-scale weather influences cloud formation. Only in the last decade have it been realised that the formation and the life cycle of the sub micron aerosol can also be driven by large-scale meteorological systems. The first such system to be identified was at low latitudes the Hadley cell, which drives aerosol formation by the transport of precursor gases in the Intertropical Convergence Zone (ITCZ) from the boundary layer to the upper troposphere, where aerosol nucleation is more efficient due to lower temperatures (e.g. Raes et al., 2000): The aerosol is assumed to be transported to the boundary layer in the subsidence regions at ~30° N and S of the equator by entrainment. In the current study, we believe we have found a similar link between the mid to high-latitude large-scale weather systems and the aerosol formation and life cycle.

ACPD
6, 10425–10462, 2006

4.3 Potential climate feed back

The new nucleated particles are often observed to grow to CCN size in 1–2 days (e.g. Kulmala et al., 2001a), fast enough for the nucleation to change the CCN population within the lifetime of the same weather system. Even though the morning hours are usually cloud free on aerosol formation days, fair weather Cumulus clouds often form in the afternoon. It may even be possible that aerosol particles formed during an early stage of a cold air outbreak over Europe have a short term feed back through the influence on cloud albedo and radiative forcing during the late stage of the same cold air outbreak, see Fig. 11. That would act to preserve the cold characteristic of the Arctic air somewhat longer during its transport southward. This possible effect of aerosol formation on CCN concentration and cloud albedo are of special interest since it is connected to the lateral transport of cold air from the Arctic to lower latitudes.

We must also consider a possible long-term climate effect. Most general circulation models (GCM) simulations of the climate effects of a doubled carbon dioxide (CO_2) concentration predicts larger temperature increases for high latitudes than for lower latitudes, and a reduction of the central Arctic pack ice (IPCC, 2001). Indeed, such a reduction in ice cover has been observed in recent years (e.g. Johannessen at al., 1999). This would affect the formation region for the Arctic air masses considerably and therefore decrease the frequency and change the characteristics of the Arctic air masses. If thereby the formation of CCN by nucleation and growth would be less frequent over Europe, this could constitute a positive feed back on the climate change.

A change in the latitudinal temperature gradients would probably change also the cyclone activity and the latitudinal exchange of air masses. Furthermore, changes in anthropogenic or natural emissions of precursor gases in northern Europe in the southbound Arctic air masses could also alter the CCN formation. This applies also to changes in primary emissions of aerosol particles. Reduced emissions of anthropogenic aerosol particles may increase secondary aerosol formation and cause a feed back effect through the net formation of CCN, which has been shown recently by

ACPD
6, 10425–10462, 2006

Spracklen et al. (2006). In future the magnitude of these potential feedback processes should be investigated in detail.

5 Summary and conclusions

The analysis of 54 months of observations of aerosol size spectra considering the occurrence of aerosol formation and growth together with air mass analysis and supporting data representing precursor sources and sinks have lead us to several interesting conclusions:

1. We have been able to generalise the results of the BIOFOR project (Nilsson et al., 2001b): aerosol formation is most frequent in Arctic (A) air, and to some degree in sub-Polar (P) air masses. Almost every second day with aerosol formation has an Arctic air mass (49%), and almost one fourth of the days with Arctic air have aerosol formation (23%). Both are approximately twice as high frequencies as the second air mass type (Polar air with 28 and 11%, respectively) and even higher than the other air mass types. In particularly, the outbreak of (usually marine) Arctic air over continental northern Europe causes the most favourable combined conditions of air in transition (x) for aerosol formation regarding photochemistry, precursor sources and sinks and boundary layer dynamics (see Fig. 11). There is however aerosol formation events in every air mass type except Subtropical (S) air, however much less frequent.

2. In each air mass type a different set of key factors favour aerosol formation:

   (a) The key factors that generally favour aerosol formation in Arctic air appears to be lower pre-existing aerosol surface (and therefore less condensational sink, CS) due to its origin in the far north remote from most anthropogenic sources. Relatively high concentration of SO_2, probably mainly from regional and local sources, may also favour the aerosol formation. Low UV-B radiation

appear to be the limiting factor that prevent aerosol formation in some days, see Table 3. Hence, clear skies and high radiation is in our analysis the factors that most critically determines if a day with Arctic air becomes an aerosol formation day.

(b) Polar air has not as low CS as A air, see Table 2, but still low UV-B, see Table 1. We can see the same sensitivity to the UV-B level as for A air in that it is the radiation that limits the possibility for aerosol formation. What is then the factor that makes sub-Polar air the second most favourable air mass for aerosol formation? If we consider the difference in geographical origin of the A and P air (compare Figs. 8 and 9 in Tunved et al., 2004), we realise that while A air has an origin north of the Scandinavian peninsula over the Arctic ocean, the sub-Polar air roughly originates from within the northern Europe. Hence, it have spent longer time over the Boreal forest and will have received larger amounts of organic emissions that may contribute to the aerosol formation. This is a possible explanation, which we cannot test since long term measurements of such emissions are not available.

(c) Heated Polar air (Ps) has generally unfavourable conditions with high CS, which explain the low frequency of aerosol formation. When aerosol formation occurs in this air mass, it is typically due to a strong increase in SO_2, most likely anthropogenic, see Table 3.

(d) Mid-latitude air (Sp) has also high CS levels and hence low aerosol formation frequency. Despite this, aerosol formation occasionally takes place thanks to large reductions in CS and/or increases in SO_2 concentration (on average doubled SO_2 concentrations), see Table 3.

(e) Low CS also favours aerosol formation when there is a marine character on the air mass, but not in mixed or continental air. Although low SO_2 concentrations are a limiting factor for aerosol formation in marine air (the lowest average level, see Table 2), it is still reductions in the CS that most likely

determine on which day aerosol formation is initiated, see Table 3.

(f) Continental air has a high SO_2 level, but have despite that only very few aerosol formation events. This is most likely because of high CS and low UV-B, which are caused by high pollution levels and high cloudiness, respectively. If the skies clear up, radiation may cause enough precursor production to induce aerosol formation despite the CS level, see Table 3.

(g) Despite that a mixed air characteristic are more favourable for aerosol formation than marine and continental air, we find no clear cause in our study, unless a combination of relatively low CS and relatively high SO_2 concentration (a compromise between the marine and arctic air) is enough. Mixed air is in our data also often the same as air in transition from mA towards cA or cP air. Hence, it constitutes air masses that initially are relatively cold due to their origin over the Arctic Ocean, which will experience gradual heating from below during the transport. This will destabilise the air and should on average cause a more turbulent and convective boundary layer that may favour aerosol formation (e.g. Nilsson et al., 2001b; Lauros et al., 2006; Hellmuth, 2006). It may be that the explanation is the same as we speculate above for P air: large volatile organic emissions from the Boreal forest once the previously marine enters the north European continent. CS is the critical condition that decides if a day with mixed air has aerosol formation or not, see Table 3.

One overall valid conclusion is that the positive effect of low CS on aerosol formation shown in several studies (e.g. Hyvönen et al., 2005; Sogacheva et al., 2005) does not apply to all air masses: sub-Polar air masses and continental air is excluded from this rule.

3. The strong annual cycle in the presence of A air (especially in Arctic air in transition from marine to continental type (xA)) with a minima in summer, is together with an annual cycle similar to that of UV-B able to explain the annual cycle in

aerosol formation frequency, and large part of the month-to-month and inter-annual variability.

4. This suggests that the monthly probability of aerosol formation is a combination of the synoptic weather and air mass type (and characteristics of the air), and either the biologically driven emissions of precursors, or the photo-chemically driven production of precursors.

This does not exclude that other major factors are not involved, for which no continuous measurements are available. We know from additional measurements during intense field campaigns that higher concentrations of NH_3 favour aerosol formation (Janson et al., 2001; Kulmala et al., 2001a), and higher concentration of oxidation products of terpenes will favour the initial steps of the growth from 1 nm to 3 nm (Kulmala et al., 2004c). Also entrainment of air with low aerosol concentration and/or high precursor concentrations could favour aerosol formation and the boundary layer dynamics itself may also enhance aerosol formation (Nilsson et al., 2001a). Non of these factors (ammonia, organics and boundary layer dynamics) have been examined in our study, due to lack of relevant long term continuous measurements.

We have finally pointed out that the cyclones that pass over northern Europe has a key role for the aerosol life cycle in more than one way, that this system and its effects on the aerosol and its air quality and climate influence and feed back effects must be considered in its whole. It may seam far-reaching to consider this as a large-scale weather/aerosol life cycle system, and unrealistic to ever consider the ability to study the whole system. However, recent observations over central Europe demonstrates that on rare occasions one may even bee able to grasp the whole cycle even without large-scale models (Heintzenberg et al., 2003). Heintzenberg manages to follow an air mass from Europe into the Arctic and back to central Europe, and because there is no new aerosol formation in this particularly case, the Arctic Haze properties are preserved, which simplifies the task to prove its origin. We believe that in most cases the Arctic air mass will experience considerably new aerosol sources, primary and/or

ACPD
6, 10425–10462, 2006

secondary, that will transform the original aerosol, and complicate the picture. We must therefore agree with Heintzenberg et al. that it will be absolutely necessary to include detailed aerosol schemes in large-scale models if we are to understand the aerosol life cycles, as well as the aerosol climate effects. Such studies are still rare, but Spracklen et al. (2006) has managed to simulate Hyytiälä type aerosol formation in a large scale model, and having done so, they demonstrated a possible feed back effect between anthropogenic and natural, primary and secondary, aerosol sources in this weather system.

Acknowledgements. Finally, we would like to acknowledge the financial support from the European commission through the QUEST project, the Swedish Research Council, and the Nordic Centre of Excellence BACCI. We also received valuable help from B. Nordman, M. Boy, and M. dal Maso when constructing the database we have used, and many more contributed to the five years of Hyytiälä measurements.

References

Aalto, P., Hämeri, K., Becker, E., Weber, R., Salm, J., Mäkelä , J. M., Hoell, C., O'Dowd, C. D., Karlsson, H., Hansson, H.-C., Väkevä, M., Koponen, I., Buzorius, G., and Kulmala, M.: Physical characterization of aerosol particles during nucleation events, logical and gas phase parameter influence, Tellus, 53B, 344–358, 2001.

Birmili, W. and Wiedensohler, A.: New particle formation in the continental boundary layer: Meteorological and gas phase parameter influence, Geophys. Res. Lett., 27, 3325–3328, 2000.

Buzorius, G., McNaughton, C. S., Clarke, A. D., Covert, D. S., Blomqvist, B., Nielsen, K., and Brechtel, F. J.: Secondary aerosol formation in continental outflow oonditions during ACE-Asia, J. Geophys. Res., 109, D24203, doi:10.1029/2004JD004749, 2004.

Coe, H., Williams, P. I., McFiggans, G., Gallagher, M. W., Beswick, K. M., Bower, K. N., and Choularton, T. W.: Behaviour of ultrafine particles in continental and marine air masses at a rural site in the United Kingdom, J. Geophys. Res., 105, 26 891–26 905, 2000.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen,

K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, Bor. Environ. Res., 10, 323–336, 2005.
Gaydos, T. M., Stanier, C. O., and Pandis, S. N.: Modeling of in situ ultrafine atmospheric particle formation in the eastern United States, J. Geophys. Res., 110, D07S12, doi:10.1029/2004JD004683, 2005.
Heintzenberg, J. and Larssen, S.: SO_2 and SO_4 in the Arcitc: interpretation of observations at three Norwegian Arcitc-subArctic stations, Tellus, 35B, 255–265, 1983.
Heintzenberg, J., Tuch, T., Wehner, B., Wiedensohler, A., Wex, H., Ansmann, A., Mattis, I., Müller, D., Wendisch, M., Eckhardt, S., and Stohl, A.: Arctic Haze over Central Europe, Tellus, 55B, 796–807, 2003.
Held, A., Nowak, A., Birmilli, W., Wiedenshohler, A., Forkel, R., and Klemm, O.: Observations of particle formation and growth in a mountainous forest region in central Europe, J. Geophys. Res., 109, D23204, doi:10.1029/2004JD005346, 2004.
Hellmuth, O.: Columnar modelling of nucleation burst evolution in the convective boundary layer – first results from a feasibility study Part III: Preliminary results on physicochemical model performance using two “clean air mass” reference scenarios, Atmos. Chem. Phys., 6, 4231–4251, 2006a.
Hellmuth, O.: Columnar modelling of nucleation burst evolution in the convective boundary layer – first results from a feasibility study Part IV: A compilation of previous observations for valuation of simulation results from a columnar modelling study, Atmos. Chem. Phys., 6, 4253–4274, 2006b.
Hyvönen, S., Junninen, H., Laakso, L., Dal Maso, M., Grönholm, T., Bonn, B., Keronen, P., Aalto, P., Hiltunen, V., Pohja, T., Launiainen, S., Hari, P., Mannila, H., and Kulmala, M.: A look at aerosol formation using data mining techniques, Atmos. Chem. Phys., 5, 3345–3356, 2005.
Institute für Meteorologie: Berliner Wetterkarte, Freie Univ. Berlin, Germany, 1997.
IPCC, Climate Change 2001: The Scientific Basis, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., and Xiaosu, D., Cambridge Univ. Press, UK, 2001.
Janson, R., Rosman, K., Karlsson, A., and Hansson, H.-C.: Biogenic emissions and gaseous precursors to Characteristics of the aerosol particle formation events the forest aerosol, Tellus, 53B, 423–440, 2001.
Johannessen, O. M., Shalina, E. V., and Miles, M. W.: Satellite evidence for an Arctic sea ice cover in transformation, Science, 286, 1937–1939, 1999.

Kulmala, M., Hämeri, K., Aalto, P. P., Mäkelä, J. M., Pirjola, L., Nilsson, E. D., Buzorius, G., Rannik, Ü, Dal Maso, M., Seidl, W., Hoffmann, T., Jansson, R., Hansson, H.-C., Viisanen, Y., Laaksonen, A., and O'Dowd, C. D.: Overview of the international project on Biogenic aerosol formation in the boreal forest (BIOFOR), Tellus, 53B, 324–343, 2001a.

Kulmala, M., Dal Maso, M., Mäkelä, J. M., Pirjola, L., Väkevä, M., Aalto, P., Miikkulainen, P., Hämeri, K., and O'Dowd, C. D.: On the formation, growth and composition of nucleation mode particles, Tellus, 255, 423–430, 2001b.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: A review of observations, J. Aerosol Sci., 35, 143–176, 2004a.

Kulmala, M., Suni, T., Lehtinen, K.E.J., Dal Maso, M., Boy, M., Reissell, A., Rannik, Ü., Aalto, P., Keronen, P., Hakola, H., Bäck, J., Hoffmann, T., Vesala, T., and Hari, P.: A new feedback mechanism linking forests, aerosols and climate, Atmos. Chem. Phys., 4, 557–562, 2004b.

Kulmala, M., Kerminen, V.-M., Anttila, T., Laaksonen, A., and O'Dowd, C. D.: Organic aerosol formation via sulphate cluster activation, J. Geophys. Res., 109, D04205, doi:10.1029/2003JD003961, 2004c.

Kulmala, M., Lehtinen, K. E. J., and Laaksonen, A.: Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration, Atmos. Chem. Phys., 6, 787–793, 2006.

Lauros, J., Nilsson, E. D., Vehkamäki, H., and Kulmala, M.: Atmospheric variability and binary homogeneous nucleation: A parametrisation and conditions required for a significant effect, J. Atmos. Res., in press, 2006.

McNaughton, C. S., Clarke, A. D., Howell, S. G., Moore II, K. G., Brekhovskikh, V., Weber, R., Orsini, D. A., Covert, D., Buzorius, G., Brechtel, F. J., Carmichael, G. R., Tang, Y., Eisele, F. L., Mauldin, R. L., Bandy, A. R., Thornton, D. C., and Blomqvist, B.: Spatial distribution and size evolution of particles in Asian outflow: Significance of primary and secondary aerosols during ACE-Asia and Trace-P, J. Geophys. Res., 109, D19S06, doi:10.1029/2003JD003528, 2004.

Mäkelä, J. M., Aalto, P., Jokinen, V., Pohja, T., Nissinen, A., Palmroth, S., Markkanen, T., Seitsonen, K., Lihavainen, H., and Kulmala, M.: Observations of ultrafine aerosol particle formation and growth in boreal forest, Geophys. Res. Lett., 24, 1219–1222, 1997.

Mäkelä, J. M., Dal Maso, M., Pirjola, L., Keronen, P., Laakso, L., Kulmala, M., and Laaksonen, A.: Characteristics of the atmospheric particle formation events observed at a boreal forest

site in sothern Finland, Boreal Environ. Res., 5, 299–313, 2000.
Mönkkönen, P., Koponen, I. K., Lehtinen, K. E. J., Hämeri, K., Uma, R., and Kulmala, M.: Measurements in a highly polluted Asian mega city: Observations of aerosol number size distribution, modal parameters and nucleation events, Atmos. Chem. Phys., 5, 57–66, 2005.
Nilsson, E. D. and Kulmala, M.: The potential for atmospheric mixing processes to enhance the binary nucleation rate, J. Geophys. Res., 103, 1381–1389, 1998.
Nilsson, E. D., Pirjola, L., and Kulmala, M.: The effect of atmospheric waves on aerosol nucleation and size distribution, J. Geophys. Res., 105, 19 917–19 926, 2000.
Nilsson, E. D., Rannik, Ü., Kulmala, M., Buzorius, G., and O'Dowd, C.: Effects of continental boundary layer evolution, convection, turbulence and entrainment on aerosol formation, Tellus, 53B, 441–461, 2001a.
Nilsson, E. D., Paatero, J., and Boy, M.: Effects of air masses and synoptic weather on aerosol formation in the continental boundary layer, Tellus, 53B, 462–478, 2001b.
Nilsson, E. D. and Leck, C.: A pseudo-Lagrangian study of the Sulfur Budget in the remote Arctic Marine Boundary Layer, Tellus, 54B, 213–230, 2002.
Raes, F., van Dingenen, R., Vignati, E., Wilson, J., Putaud, J.-P., Seinfeld, J. H., and Adams, P.: Formation and cycling of aerosols in the global troposphere, Atmos. Environ., 34, 4215–4240, 2000.
Rahn, K. A.: Elemental tracers for source regions of Artic pollution aerosol, Idöjaras, 86, 1–14, 1982.
Rodriguez, S., Van Dingenen, R., Putaud, J.-P., Martins-Dos Santos, S., and Roselli, D.: Nucleation and growth of new particles in the rural atmosphere of Northern Italy – relationships to air quality monitoring, Atmos. Environ., 39, 6734–6746, 2005.
Sogacheva L., Dal Maso, M., Kerminen, V.-M., and Kulmala, M.: Probability of nucleation events and aerosol particle concentration in different air mass types arriving at Hyytiäkä, Southern Finalnd, based on back trajectories, Boreal Environ. Res., 10, 479–491, 2005.
Spracklen, D. V., Carslaw, K. S., Kulmala, M. , Kerminen, V.-M., Mann, G. W., and Sihto, S.-L.: The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales, Atmos. Chem. Phys. Discuss., 6, 7323–7368, 2006.
Stratmann F., Siebert, H. , Spindler, G., Wehner, B., Althausen, D., Heintzenberg, J., Hellmuth, O., Rinke, R., Schmieder, U., Seidel, C., Tuch, T., Uhrner, U., Wiedensohler, A., Wandinger, U., Wendisch, M., Schell, D., and Stohl, A.: New-particle formation events in a continental boundary layer: first results from the SATURN experiment, Atmos. Chem. Phys., 3, 1445–

1459, 2003.
Tunved, P., Nilsson, E. D., Hansson, H.-C., and Ström, J.: Aerosol characteristics of air masses in Northern Europe- influence of location, transport, sinks and sources, J. Geophys. Res., 110, 7201, doi:10.1029/2004JD005085, 2005.
Uhrner, U., Birmili, W., Stratmann, F., Wilck, M., Ackermann, I. J., and Berresheim, H.: Particle formation at a continental background site: comparison of model results with observations, Atmos. Chem. Phys., 3, 347–359, 2003.
Vana, M., Kulmala, M., Dal Maso, M., Hörrak, U., and Tamm, E.: Comparative study of nucleation mode aerosol particles and intermediate air ions formation events at three sites, J. Geophys. Res., 109, doi:10.1029/2003JD004413, 2004.
Väkevä, M., Hämeri, K., Puhakka, T., Nilsson, E. D., Hohti, H., and Mäkelä, J. M.: Effects of meteorological processes on aerosol particles size distribution in an urban background area, J. Geophys. Res., 105, 9807–9821, 2000.

Table 1. Characteristics of air masses with and without aerosol formation.

	SO_2 (ppb)	UV-B (Wm^{-2})$_{\#\$}$	CS (s^{-1})	CS (s^{-1})$_{\#}$	CS (s^{-1})$_{\%}$
			All days		
Arctic (A)	0.61	0.30	3.4×10^{-3}	2.2×10^{-3}	2.3×10^{-3}
sub-Polar (P)	0.54	0.33	3.8×10^{-3}	3.6×10^{-3}	3.9×10^{-3}
heated sub-Polar (Ps)	0.50	0.58	3.3×10^{-3}	5.0×10^{-3}	5.3×10^{-3}
mid-latitude (Sp)	0.45	0.57	4.7×10^{-3}	6.3×10^{-3}	7.3×10^{-3}
sub-tropical (S)	0.57	0.87	No data	11.2×10^{-3}	13.0×10^{-3}
marine (m)	0.25	0.32	3.9×10^{-3}	2.9×10^{-3}	3.1×10^{-3}
transition (x)	0.56	0.45	3.5×10^{-3}	3.7×10^{-3}	4.0×10^{-3}
continental (c)	1.53	0.18	3.3×10^{-3}	4.6×10^{-3}	5.0×10^{-3}
			Aerosol formation days relative to all days (for the given air mass)		
Arctic (A)	−30%	+40%	−3%	−23%	−9%
sub-Polar (P)	−4%	+42%	+26%	−8%	−8%
heated sub-Polar (Ps)	+54%	−10%	+27%	−22%	−19%
mid-latitude (Sp)	+96%	+12%	+51%	−62%	−70%
sub-tropical (S)	−14%	−56%	No data	No data	No data
Marine (m)	−12%	+16%	−15%	−45%	−32%
Transient (x)	+2%	+11%	+14%	−35%	−30%
Continental (c)	−10%	+83%	+24%	+2%	+2%

$_{\#}$ 09:00–12:00 h local time
$_{\%}$ 06:00–09:00 h local time
$_{\$}$ only from April 1997 to December 1999

ACPD
6, 10425–10462, 2006

Table 2. Condensational sink (CS) in different air masses with and without aerosol formation.

	CS (s^{-1})	CS $(s^{-1})_{\#}$	CS $(s^{-1})_{\%}$
	Days with aerosol formation		
Arctic (A)	3.3×10^{-3}	1.7×10^{-3}	2.1×10^{-3}
sub-Polar (P)	4.8×10^{-3}	3.3×10^{-3}	3.6×10^{-3}
heated sub-Polar (Ps)	4.2×10^{-3}	3.9×10^{-3}	4.3×10^{-3}
mid-latitude (Sp)	7.1×10^{-3}	2.4×10^{-3}	2.2×10^{-3}
sub-tropical (S)	No data	No data	No data
Marine (m)	3.3×10^{-3}	1.6×10^{-3}	2.1×10^{-3}
Transient (x)	4.0×10^{-3}	2.4×10^{-3}	2.8×10^{-3}
Continental (c)	4.1×10^{-3}	4.7×10^{-3}	5.1×10^{-3}
	Days without aerosol formation		
Arctic (A)	3.4×10^{-3}	2.3×10^{-3}	2.3×10^{-3}
sub-Polar (P)	3.7×10^{-3}	3.7×10^{-3}	3.9×10^{-3}
heated sub-Polar (Ps)	3.2×10^{-3}	5.1×10^{-3}	5.5×10^{-3}
mid-latitude (Sp)	4.4×10^{-3}	6.3×10^{-3}	7.2×10^{-3}
Sub-tropical (S)	No data	11.2×10^{-3}	13.0×10^{-3}
Marine (m)	4.0×10^{-3}	3.1×10^{-3}	3.4×10^{-3}
Transient (x)	3.4×10^{-3}	3.9×10^{-3}	4.1×10^{-3}
Continental (c)	3.2×10^{-3}	4.6×10^{-3}	4.9×10^{-3}

$_{\#}$ 09–12:00 h local time
$_{\%}$ 06–09:00 h local time

ACPD
6, 10425–10462, 2006

Table 3. Summary of factors that favour aerosol formation in each air mass.

	Aerosol formation frequency	General conditions		Critical conditions
		positive	negative	
Arctic (A)	49%	low CS	low UV-B	+40% UV-B
sub-Polar (P)	28%	*high OC emissions?*	low UV-B	+40% UV-B
heated sub-Polar (Ps)	6%		high CS	+50% SO_2
mid-latitude (Sp)	3%		high CS	+100% SO_2 -70% CS
Sub-tropical (S)	0%	high UV-B	high CS	no data
Marine (m)	31%	low CS	low SO_2	−40% CS
Transient (x)	48%	*Unstable BL?*		−40% CS
Continental (c)	9%	high SO_2	low UV-B high CS	+80% UV-B

Italic indicates a speculation rather than a result of our data analysis. OC=Organic Carbon, BL=Boundary Layer.

ACPD
6, 10425–10462, 2006

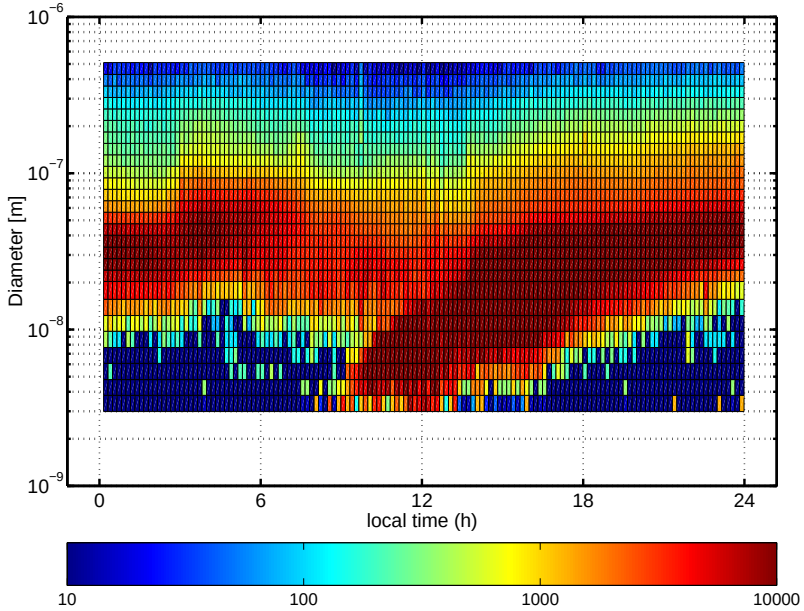


Fig. 1. Example of the evolution of the aerosol number size distribution during a first class aerosol formation day (10 April 1998) in Hyytiälä, measured with a DMPS-system. The color scale indicates the aerosol number concentration per size increment and size scan.

ACPD
6, 10425–10462, 2006

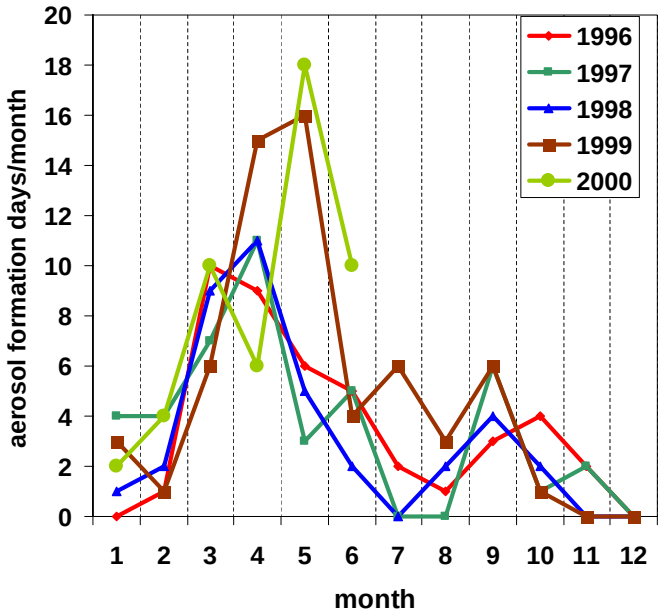


Fig. 2. Monthly number of aerosol formation days for the years 1996 to 2000.

ACPD
6, 10425–10462, 2006

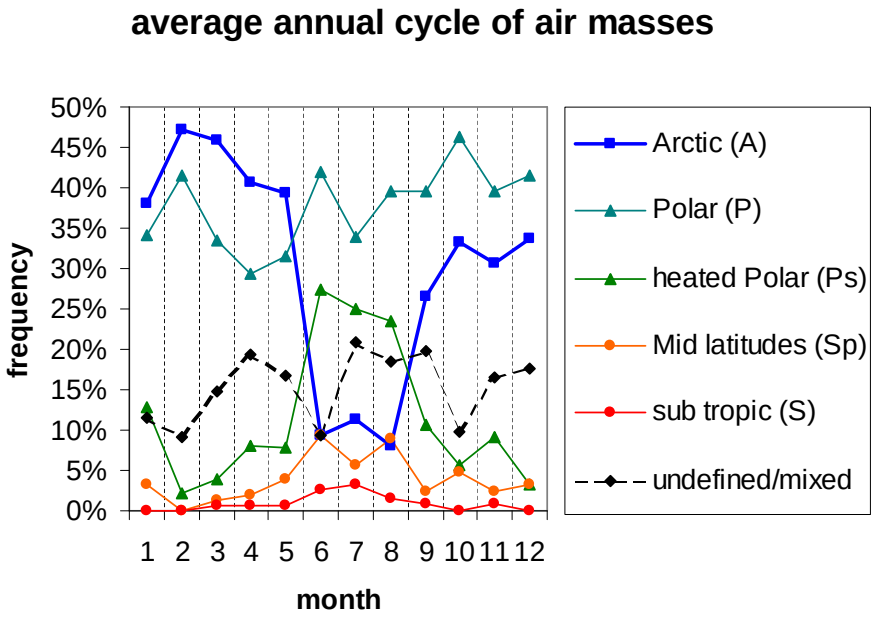


Fig. 3. Monthly frequency of different air masses over Hyytiälää, averaged for 1996 to 2000.

ACPD
6, 10425–10462, 2006

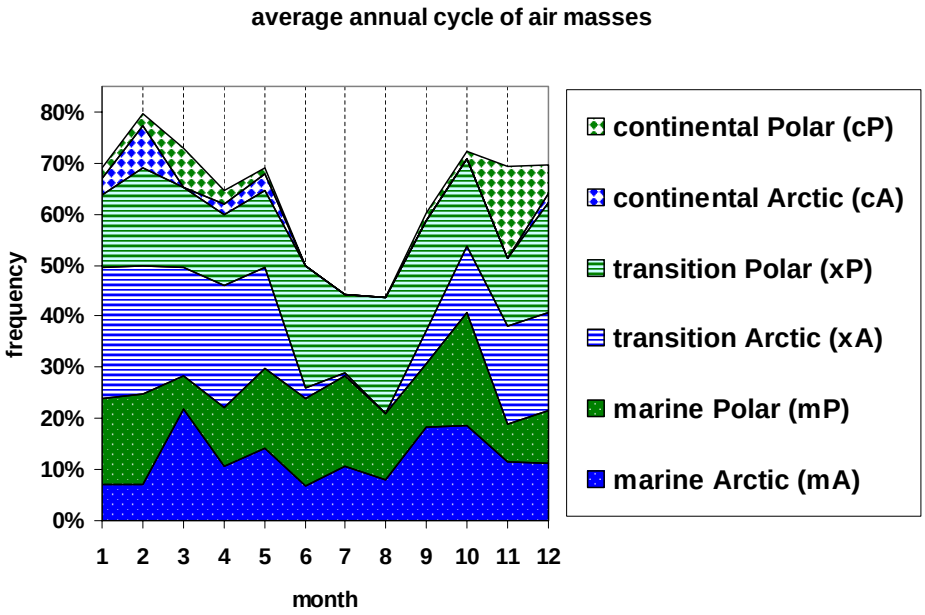


Fig. 4. Monthly frequency of the marine, transient or continental characteristic of the sub-Polar and Arctic air masses over Hyytiälää, averaged for 1996 to 2000.

ACPD
6, 10425–10462, 2006

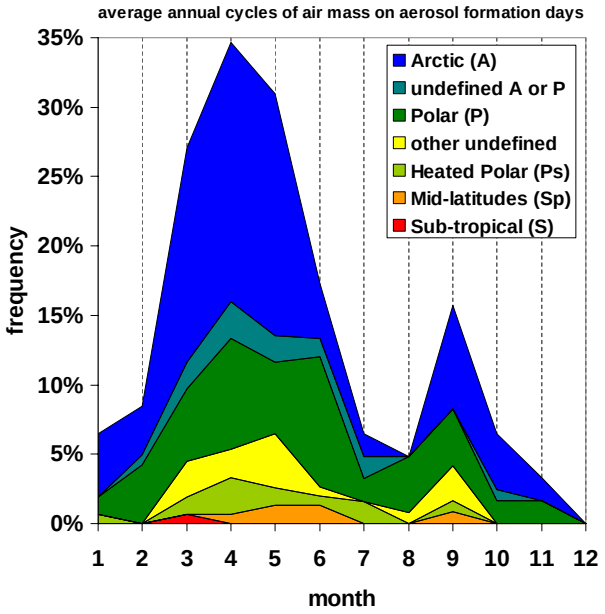


Fig. 5. Monthly frequency of different air masses over Hyytiälää during aerosol formation days, averaged for 1996 to 2000.

ACPD
6, 10425–10462, 2006

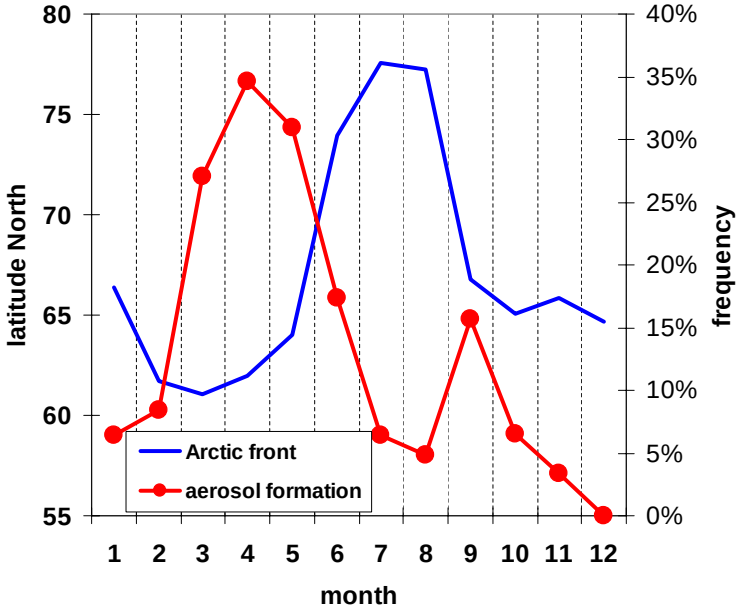


Fig. 6. Monthly average location of the Arctic and Polar fronts along the 15° E longitude, and the monthly frequency of days with aerosol formation, averaged for 1996 to 2000.

ACPD
6, 10425–10462, 2006

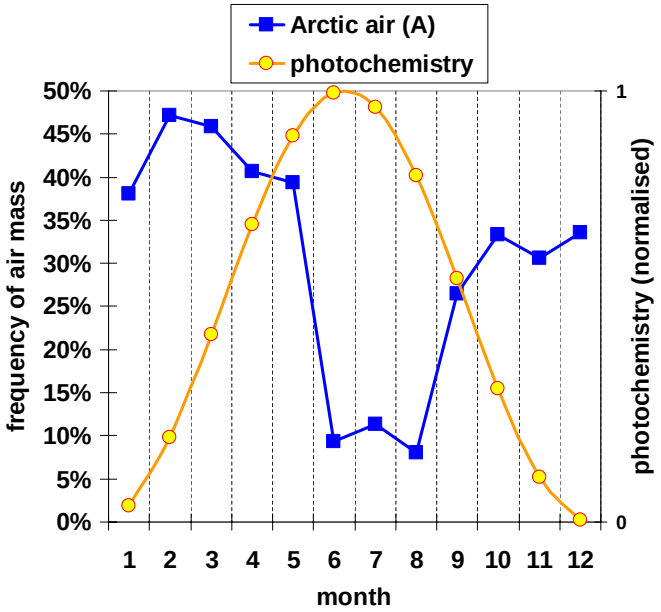


Fig. 7. Monthly frequency for Arctic air over Hyytiälä, averaged over the period 1996 to 2000, and a non-dimensional proxy curve for the photochemistry with the same phase and approximate shape as the annual UV-B cycle.

ACPD
6, 10425–10462, 2006

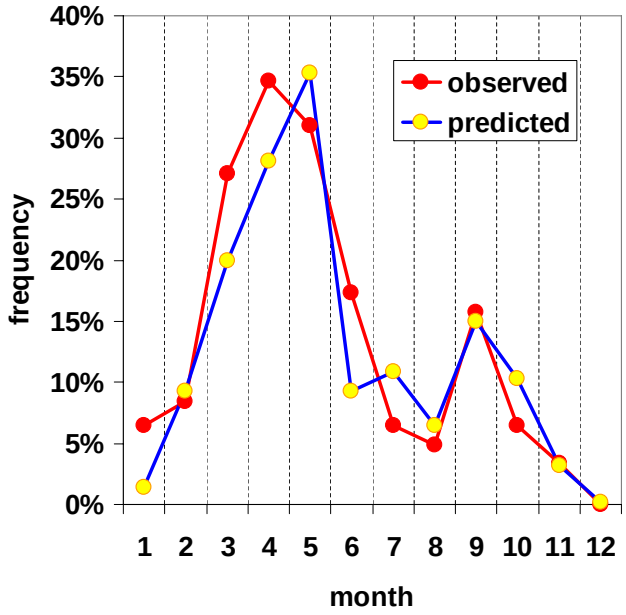


Fig. 8. Monthly frequency of observed and predicted aerosol formation days in Hyytiälä, averaged over the period 1996 to 2000.

ACPD
6, 10425–10462, 2006

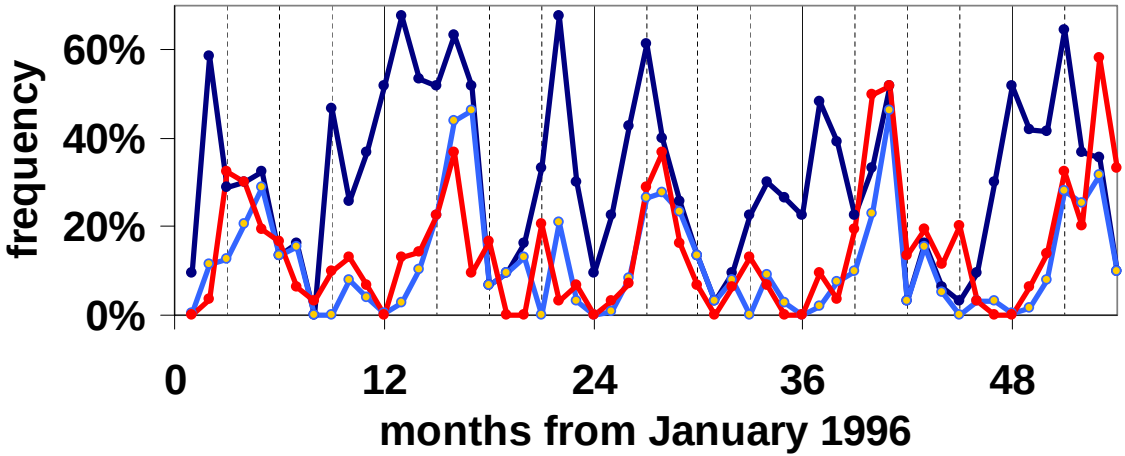


Fig. 9. Frequencies of air masses and aerosol formation month by month from January 1996, Hyytiälä, Finland. Frequency of Arctic air masses month by month (dark blue line and circles). Observed aerosol formation frequency (red line and circles). Aerosol formation frequency predicted from the Arctic air mass frequency and the probability of photo chemically driven aerosol formation (light blue line and yellow circles).

ACPD
6, 10425–10462, 2006

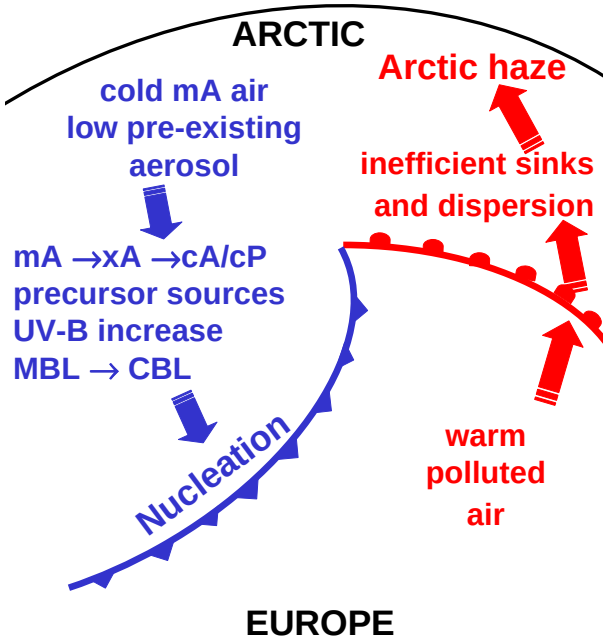


Fig. 10. Schematic figure of the effects of a cyclone passage over northern Europe on the aerosol formation during the spring. In each cyclone system there is a warm polluted air mass advancing behind a warm front (red line) and a cold clean air mass advancing behind a cold front (blue line). The result is often transport of warm polluted air from Europe to the Arctic, which contributes to the Arctic Haze, and cold and clean Arctic air to Europe, which favour aerosol formation.

ACPD
6, 10425–10462, 2006

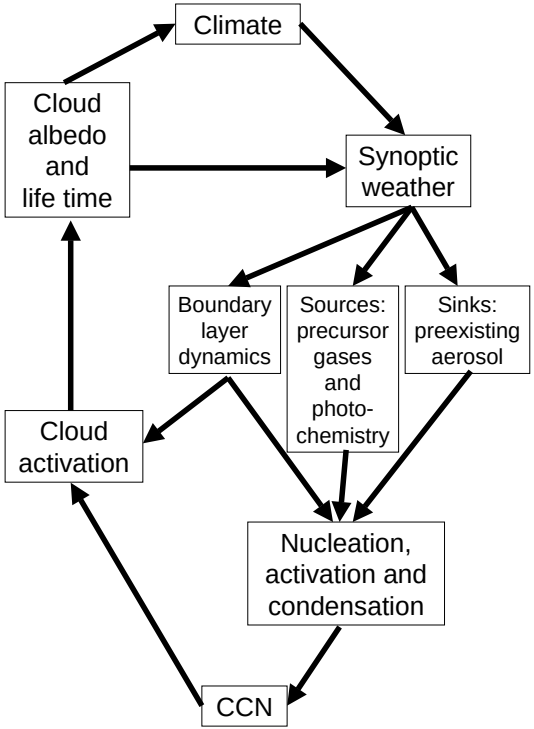


Fig. 11. Schematic figure of the weather-aerosol formation-climate-weather feedback. Synoptic weather features such as frequencies of Arctic air, cyclone activity or paths are dependent on climate and affects on **(a)** boundary layer dynamics, **(b)** gas phase chemistry and **(c)** concentration of pre-existing aerosols. All of (a), (b) and (c) are key factors in aerosol formation and growth. After growth a fraction of the aerosols form CCN, which together with trace gases and boundary layer dynamics affect cloud formation. The number of cloud droplets affect on cloud albedo and lifetime, which causes a feedback to both the synoptic weather and climate.

ACPD
6, 10425–10462, 2006